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(54) Silicone gel composition

Silikongelzusammensetzung Composition de gel de silicone

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(73) Proprietor: Dow Corning Toray Silicone Company, Limited Tokyo 103 (JP)

(72) Inventor: Harashima, Asao Ichihara-shi, Chiba Prefecture (JP)

(74) Representative: Spott, Gottfried, Dr. Spott, Weinmiller & Partner Sendlinger-Tor-Platz 11 80336 München (DE)

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EP-A- 0 076 146 EP-A- 0 407 089 EP-A- 0 456 459

EP-A- 0 298 402

EP-A- 0 435 483

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#### Remarks:

The file contains technical information submitted after the application was filed and not included in this specification

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### Description

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The present invention relates to a silicone gel composition and, more specifically, relates to a silicone gel composition that gives a light, dry sensation or feel and that is very stable with respect to time.

Silicone oils are nontoxic, highly spreadable, highly lubricating, and very water repellent, and for these reasons are employed in such products as cosmetics, makeup, skin-care cosmetics, hair-care cosmetics, and pharmaceutical products. Silicone oils have been available in the form of water-based silicone emulsion compositions, silicone oil compositions, and silicone gel compositions. Silicone gel compositions have been used for sunscreen gels, moisturizing creams, antiperspirant creams, liquid foundations, and hair gels.

Known silicone gel compositions include silicone gel compositions composed of silicone oil and wax and silicone gel compositions composed of silicone oil and silica or lipophilic silica. However, it is difficult to smoothly spread the wax-containing silicone gel compositions on the skin or hair, and once applied these compositions give a clammy feel. On the other hand, the silica-containing silicone gel compositions have poor keeping qualities because the silica aggregates and the silicone oil separate out. In response to this, Japanese Patent Application Laid Open [Kokai or Unexamined] Number Sho 61-113646 [113,646/1986] has proposed a silicone gel composition that consists of silicone oil, a polyoxyalkylene-containing organopolysiloxane, an organically modified clay mineral, such as dioctadecyldimethylammonium salt-modified montmorillonite, and water. Nonetheless, the compositions of Sho 61-113646 are still problematic. Furthermore, this silicone gel composition still provides a clammy feel when applied to the skin or hair.

In Japanese patent applications JP-A 61 212 324, JP-A 62 045 656, JP-A 62 054 759 and JP-A 61 113 646 silicone oil containing gel compositions are disclosed which comprise a silicone oil, a polyoxyalkylene modified organopolysiloxane and as essential ingredient an organically modified clay mineral together with water. The polyoxyalkylene modified organopolysiloxane used in these compositions can be a compound which has polyoxyalkylene groups at the molecular ends of the modified organopolysiloxane. The molecules contain only one sort of polyoxyalkylene groups, either oxyethylene groups or oxypropylene groups.

EP-A 0 076 146 discloses skin conditioning compositions which are in the form of an emulsion. The emulsion comprises a high amount of glycerine, 0,5 to 10 percent of a volatile silicone, 0,05 to 1 percent of a polydiorganosiloxane polyalkylene copolymer which contains at least one polyoxyalkylene segment, a surfactant and water. The description of the polydiorganosiloxane polyalkylene copolymer is very general. In a preferred embodiment the copolymer comprises dimethylsiloxane units and methylsiloxane units which carry polyoxyalkylene segments and as terminating groups trimethylsilyl groups.

From EP-A 0 456 459, EP-A 0 435 483, EP-A 0 407 089 and EP-A 0 298 402 it is known to use a polymer of dimethylpolysiloxane with polyoxyethylene and/or polyoxypropylene side chains as surfactant for a water in silicone oil emulsion wherein the emulsion further comprises a volatile polydimethylsiloxane and other ingredients which are useful for such a product.

The present invention takes as its object the introduction of a silicone gel composition that gives a light, dry sensation and that is very stable with respect to time.

The present invention relates to a silicone gel composition that comprises:

(a) 20 to 95 percent by weight of a silicone oil selected from the group consisting of dimethylpolysiloxane, methylphenylpolysiloxanes, dimethylsiloxane-methylphenylsiloxane copolymers; octamethylcyclotetrasiloxane, decamethylcyclopentasiloxane, tetramethyltetraphenyltetracyclosiloxane; cyclic siloxane solutions of dimethylpolysiloxane gums, dimethylsiloxane-methylphenylsiloxane copolymer gums, and dimethylpolysiloxane gums; trimethylsiloxysilicic acids and cyclic siloxane solutions of trimethylsiloxysilicic acids; diorganopolysiloxanes having  $C_{6.50}$  alkyl groups; and amino-containing diorganopolysiloxanes;

(b) 2 to 30 percent by weight of a polyoxyalkylene organopolysiloxane with a formula selected from the group consisting of

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wherein R is the methyl or phenyl group; R' is selected from the group consisting of the hydrogen atom, an acyl group, and alkyl groups having 1 to 4 carbon atoms, <u>a</u> is an integer with a value of 5 to 50; <u>m</u> is an integer with a value of 50 to 1,000; and <u>n</u> is an integer with a value of 1 to 40; and (c) 0.2 to 80 percent by weight of water.

The silicone oil comprising component (a) is exemplified by low- to high-viscosity diorganopolysiloxanes and is selected from dimethylpolysiloxanes, methylphenylpolysiloxanes, and dimethylsiloxane-methylphenylsiloxane copolymers; cyclic siloxanes such as octamethylcyclotetrasiloxane, decamethylcyclopentasiloxane, and tetramethyltetraphenyltetracyclosiloxane; cyclic siloxane solutions of high degree-of-polymerization dimethylpolysiloxane gums, dimethylsiloxane-methylphenylsiloxane copolymer gums, and dimethylpolysiloxane gums; trimethylsiloxysilicic acids and the cyclic siloxane solutions of trimethylsiloxysilicic acids; diorganopolysiloxanes having C<sub>6-50</sub> alkyl groups; and amino-containing diorganopolysiloxanes. Component (a) can take the form of a single species of silicone oil as exemplified above or a mixture of two or more such silicone oils.

The content of component (a) in the silicone gel composition of the present invention is 20 to 95 weight %.

Component (b) functions as gelling agent in the present invention. Component (b) is a polyoxyalkylene group-containing organopolysiloxane with the general formulas as defined in claim 1.

The group R in these formulas is the methyl or phenyl group. The group R' in these polyoxyalkylene groups is the hydrogen atom, an acyl group, or an alkyl group having 1 to 4 carbon atoms. Acyl groups are exemplified by formyl, acetyl, propionyl, butyryl, acryloyl, benzoyl, and toluoyl. The  $C_{1-4}$  alkyl groups are specifically exemplified by methyl, ethyl, isopropyl, n-propyl, tert-butyl, and n-butyl. Moreover, a is an integer with a value of 5 to 50 and  $\underline{b}$  is also an integer with a value of 5 to 50. The ranges for the polyoxyalkylene group in component (b) are such that when  $\underline{a}$  or  $\underline{b}$  is also an integer with a value of 5 to 50. The ranges for the polyoxyalkylene group in component (b) are such that when  $\underline{a}$  or  $\underline{b}$  exceeds 50, the resulting silicone gel composition gives a clammy sensation. The polyoxyalkylene group content in component (b) is not specifically restricted, but the preferred polyoxyalkylene group content falls within the range of 20 to 70 weight % exclusive of 20 weight %. Component (b) has a sharply reduced thickening activity when the polyoxyalkylene group content in component (b) is  $\leq$  20 weight %. On the other hand, a content in excess of 70 weight % results in a loss of compatibility with component (a).

In addition,  $\underline{m}$  is an integer with a value of 50 to 1,000 and  $\underline{n}$  is an integer with a value of 1 to 40. The thickening activity is unsatisfactory when  $\underline{m}$  falls below 50 and  $\underline{n}$  falls below 1. On the other hand, the resulting silicone gel composition gives a clammy sensation when  $\underline{m}$  exceeds 1,000 and  $\underline{n}$  exceeds 40.

Neither the molecular weight of component (b) nor its viscosity at 25°C are specifically restricted. However, preferred viscosities for the 50 weight % octamethylcyclotetrasiloxane solution of component (b) fall within the range of 1,000 to 100,000 centistokes (mm²/sec) because this leads to the formation of a stable gel that gives a light, dry sensation.

The component (b) content is in the range of 2 to 30 weight % and its particularly preferred range is 5 to 15 weight %. A stable silicone gel composition cannot be obtained when the silicone gel composition of the present invention component (b) content exceeds 30 weight %, the silicone gel composition will give a clammy feeling.

The silicone gel composition of the present invention is prepared by mixing water into the components (a) and (b). The content of water in the silicone gel composition of the present invention preferably falls within the range of 0.2 to 80 weight % and particularly preferably falls within the range of 0.3 to 75 weight %. A stable silicone gel composition cannot be prepared when the water content falls below 0.2 weight %. When the water content exceeds 80 weight %, the water will separate from the silicone gel composition, and the preparation of a stable silicone gel composition becomes difficult.

The silicone gel composition of the present invention is storage stable, and it affords a light, dry sensation when applied to the skin or hair. These qualities make it ideal for application in cosmetics. For its cosmetic applications, the silicone gel composition of the present invention may be blended as desired with such cosmetic additives as waxes, oils and fats, lower alcohols, lower polyhydric alcohols, higher alcohols, esters, moisture-retention agents, pigments, antiperspirants, UV absorbers, fragrances, and preservatives.

Furthermore, due to its excellent keeping qualities and light, dry feel, the silicone gel composition of the present

invention can be used in pharmaceutical products, automotive polishes, and furniture polishes.

The present invention is explained in greater detail below through illustrative examples, but the present invention is not limited thereby. Table 1 shows the organopolysiloxanes used as component (b) in the examples. The structures of the organopolysiloxane referenced in Table 1 are shown below.

## Type II

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### Type III

## TABLE 1

organopolysiloxane	type	R	<u>E</u>	<u>n</u> .	<u>a</u>	Ь	R'
L	11	methyl	400	8	25	25	Н
M	- 11	methyl	400	8	25	4	Н
0	Ш	methyl	300	-	30	15	Н
Р	111	methyl	300	-	30	0	н
Q	10	methyl/phenyl = 80/20	500	-	40	60	C <sub>4</sub> H <sub>9</sub>
R	Ш	methyl/phenyl = 80/20	500	-	3	60	C <sub>4</sub> H <sub>9</sub>

The stability with respect to time was measured as follows.

### 45 Appearance

The silicone composition was sampled into a 100 cc sample bottle, and the appearance was visually inspected after standing for 30 days at room temperature.

## 50 Viscosity

Using a VDA rotary viscometer, the viscosity of the silicone gel composition was measured both immediately after preparation and again after standing for 30 days at room temperature.

### 55 Application sensation

The apreadability and sensation were evaluated when the silicone composition was applied to the skin.

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# Example 1

10 weight parts of the organopolysiloxane were dispersed into 90 weight parts of the silicone bil by stirring. Into this was mixed 1 weight part of the ultraviolet absorber Escalol 507 from Van Dyk and Company, Inc. followed by the dropwise addition of 2 weight parts water while mixing. A sunscreen gel was subsequently obtained by dispersion to homogeneity in a homomixer. These sunscreen gels were very spreadable and gave a light, dry feeling. Tables 2, and 3 show the appearance and viscosity for each product both immediately after preparation and after standing for 30 days at room temperature.

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15		Comparison Example 1	+		+	transparent	precipitate present	no separation	separated into 2 phases	15,000	9,000	5,250	1,000
20	TABLE 2	Example 1	+		+	transparent	transparent	no separation	no separation	16,000	13,000	2,750	2,700
25			ə			6				по		uo	
30			<u>silicone oil</u> decamethylpentacyclosiloxane	organopolysiloxane L M		<u>appearance</u> immediately after preparation	9)	<u>stability</u> immediately after preparation	\s	viscosity (cps) [kPa.s] rotor no. 2, 1.5 rpm immediately after preparation	S.	viscosity(cps) lkPa.sl rotor no. 3. 6 rpm inmediately after preparation	2
35			sil decamethylpe	organo		ap inmediately	after 30 days	<u>s</u> inmediately	after 30 days	Viscosity ( rotor no. immediately af	after 30 days	viscosity(cps) rotor no. 3. inmediately after	after 30 days

250 rotor no. 1 ·6 rpm 5 1.350 rotor no. 2 6 rpm 1,100 rotor no. 1 1.5 rph no separation separation Comparison Example 3 transparent transparent 2.000 잂 10 no separation no separation transparent transparent Example 6,300 5.600 2,300 2,150 15 20 rotor no. 1 30 rpm 58 rotor no. 1 30 rpm separated into 2 phases 470 rotor no. 1 1.5 rpm Comparison Example 2 cloudy white precipitate present no separation 493 rotor no. 6 rpm 25 TABLE 3 23 no separation no separation transparent transparent 30 Example 2 18,500 19,000 5,500 5,750 35 20 wt% decamethyltetra-cyclosiloxane solution of dimethylsiloxane/methyl-phenylsiloxane polyether-modified silicone viscosity (cps) (kPa.sl rotor no. 3, 6 rpm inmediately after preparation viscosity (cps) [kPa.s] inmediately after preparation appearance immediately after preparation stability inmediately after preparation 40 O A4 O4 45 after 30 days after 30 days after 30 days after 30 days copolymer

# Example 2

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4 weight parts of the organopolysiloxane were dispersed in 60 weight parts of the silicone oil by stirring. 36 weight parts 50 % aqueous aluminum chlorohydrate solution ACH303 from Dow Corning Corporation were gradually dripped in while dispersing. After completion of addition, an antiperspirant gel was obtained by mixing for 5 minutes at 3,000

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rpm in a homomixer. The resulting antiperspirant gels had a good spreadability and gave a light, dry feel. For each product Table 4 shows the appearance both immediately after preparation and after standing for 30 days at room temperature.

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	Table 4	
	Example 4	Comparison Example 4
silicone oil		
decamethylpentacyclosiloxane solution of silicone gum	+	+
<u>organopolysiloxane</u> L M	+	+
<u>appearance</u> immediately after preparation	uniform, microturbid	uniformly cloudy white
after 30 days	uniform, microturbid	nonuniform, cloudy white
stability immediately after preparation	no separation	no separation
after 30 days	no separation	separation of water
condition immediately after preparation	gel	gel
after 30 days	gel	separation

#### Claims

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### 1. A silicone gel comprising

(a) 20 to 95 percent by weight of a silicone oil selected from the group consisting of dimethylpolysiloxane, methylphenylpolysiloxanes, dimethylsiloxane-methylphenylsiloxane copolymers; octamethylcyclotetrasiloxane, decamethylcyclopentasiloxane, tetramethyltetraphenyltetracyclosiloxane; cyclic siloxane solutions of dimethylpolysiloxane gums, dimethylsiloxane-methylphenylsiloxane copolymer gums, and dimethylpolysiloxane gums; trimethylsiloxysilicic acids and cyclic siloxane solutions of trimethylsiloxysilicic acids; diorgano-polysiloxanes having C<sub>6-50</sub> alkyl groups; and amino-containing diorganopolysiloxanes;

(b) 2 to 30 percent by weight of a polyoxyalkylene organopolysiloxane with a formula selected from the group consisting of

wherein R is the methyl or phenyl group; R' is selected from the group consisting of the hydrogen atom, an acyl group, and alkyl groups having 1 to 4 carbon atoms,  $\underline{a}$  is an integer with a value of 5 to 50;  $\underline{m}$  is an integer with a value of 5 to 50;  $\underline{m}$  is an integer with a value of 50 to 1,000; and  $\underline{n}$  is an integer with a value of 1 to 40; and (c) 0.2 to 80 percent by weight of water.

## Patentansprüche

## 1. Silicongel umfassend

bestehend aus

(a) 20 bis 95 Gew.-% eines Siliconöls ausgewählt aus der Gruppe bestehend aus Dimethylpolysiloxan, Methylphenylpolysiloxanen, Dimethylsiloxan-Methylphenylsiloxan-Copolymeren; Octamethylcyclotetrasiloxai Decamethylcyclopentasiloxan, Tetramethyltetraphenyltetracyclosiloxan; cyclischen Siloxanlösungen von Dimethylpolysiloxangummis, Dimethylsiloxanmethylphenylsiloxan-Copolymergummis und Dimethylpolysiloxangummis; Trimethylsiloxykieselsäuren und cyclischen Siloxanlösungen von Trimethylsiloxykieselsäuren; Diorganopolysiloxanen mit C<sub>6-50</sub>-Alkylgruppen und aminohaltigen Diorganopolysiloxanen; (b) 2 bis 30 Gew.-% eines Polyoxyalkylenorganopolysiloxans mit einer Formel ausgewählt aus der Gruppe

worin R die Methyl- oder Phenylgruppe ist, R' ausgewählt ist aus der Gruppe bestehend aus dem Wasserstoffatom, einer Acylgruppe und Alkylgruppen mit 1 bis 4 Kohlenstoffatomen, <u>a</u> eine ganze Zahl mit einem Wert von 5 bis 50 ist, <u>b</u> eine ganze Zahl mit einem Wert von 5 bis 50 ist, <u>m</u> eine ganze Zahl mit einem Wert von 5 bis 1000 ist und <u>n</u> eine ganze Zahl mit einem Wert von 1 bis 40 ist und (c) 0,2 bis 80 Gew.-% Wasser.

#### Revendications

### 1. Gel de silicone comprenant

(a) 20 à 95 % en poids d'une huile de silicone choisie dans le groupe constitué du diméthylpolysiloxane, des méthylphénylpolysiloxanes, des copolymères diméthylsiloxane/méthylphénylsiloxane, de l'octaméthylcyclotétrasiloxane, du décaméthylcyclopentasiloxane, du tétraméthyltétraphényltétracyclosiloxane ; des solutions, dans un siloxane cyclique, de gommes de diméthylpolysiloxane, de gommes de copolymère diméthylsiloxane/méthylphénylsiloxane, et de gommes de diméthylpolysiloxane ; des acides triméthylsiloxysiliciques et des solutions, dans un siloxane cyclique, d'acides triméthylsiloxysiliciques ; des diorganopolysiloxanes ayant des groupes alkyles en  $C_6$  à  $C_{50}$ ; et des diorganopolysiloxanes contenant un groupe amino ; (b) 2 à 30 % en poids d'un polyoxyalkylène organopolysiloxane avec une formule choisie dans le groupe

(b) 2 a 30 % en poids d'un polyoxyalkylene organopolysiloxane avec une formule choisie dans le groupe constitué de

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dans lesquelles R est le groupe méthyle ou phényle; R' est choisi dans le groupe constitué de l'atome d'hydrogène, d'un groupe acyle et des groupes alkyles ayant 1 à 4 atomes de carbone, <u>a</u> est un nombre entier avec une valeur de 5 à 50, <u>b</u> est un nombre entier avec une valeur de 5 à 50; <u>m</u> est un nombre entier avec une valeur de 1 à 40; et (c) 0,2 à 80 % en poids d'eau.